Polarization transfer in ion-surface scattering

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Chapter 7

Summary and outlook

The work described in this thesis is devoted to the investigation of the possibilities of using ions as probes of the electronic structure of surfaces. One specific topic addressed was whether ion beams can be used to obtain useful information on the electron spin ordering at (polarized) surfaces. The interaction of (highly charged) ions with surfaces is dominated by electron transfer processes. Because electron spin is conserved in the electron capture processes, the polarization of the surface electrons can be transferred to the projectiles. This has has been demonstrated in the chapters 5 and 6 of this thesis. In particular for the case of multiply charged ions, neutralization takes place 'over-the-barrier' into excited projectile states which are (nearly) energetically resonant with states in the target. This leads to the formation of so-called 'hollow atoms', i.e. atoms with populated outer shells and sparsely filled inner shells. These exotic atoms decay efficiently by photon emission or by Auger transitions, which lead to electron emission. Ever since the advent of highly charged ion sources, the formation and decay of the hollow atoms has been studied extensively and by now the main mechanisms are well understood (see e.g. refs. [11] to [14]). On basis of this knowledge it is expected that the interaction of highly charged ions with a magnetic target (spin-polarized) will lead to an enhanced population of higher-spin states, as compared to the case of a non-magnetic target. In Auger Electron Spectroscopy (AES), the electron emission from different spin states can be resolved and thereby it gives access to the spin polarization of the target (chapter 5). Next to this method, Electron Capture Spectroscopy (ECS) with singly charged ions can be used. A fraction of the grazingly incident projectiles is neutralized into an excited state, which subsequently decays under photon emission (fluorescence). After the neutralization, the electron spin is via the spin-orbit coupling (partly) transferred to the orientation of the total angular momentum, and therefore linked to the polarization of the emitted light (chapter 5).

Both methods rely heavily on the knowledge of neutralization processes in ion-surface interactions. Although the electron transfer mechanisms are by and large understood, several aspects of electron transfer between surface and projectile needed further investigation. In particular, the target polarization is carried by the conduction band electrons. Therefore it is important to explore the balance between resonant capture from the conduction band and direct inner-shell capture into projectile states. The latter mechanism can be studied by a variation of the projectiles charge state and/or its velocity (chapter 4).

Therefore, in chapter 4 the interaction of slow ($v < 0.4$ a.u.) hydrogen-like ions with carbon surfaces was studied. AES was used to study the changes in the electron emission induced by the various projectiles. Surprisingly, a strong target K Auger emission was found. These KVV Auger electrons partly originate from carbon atoms at the surface and partly from those of the bulk. A second fraction of the carbon K Auger electrons exhibits distinct spectral features. These peaks can be identified as being due to atomic KLL transitions. Such strong Auger electron emission from the target has not been observed earlier. We presented strong indications that these target KLL Auger electrons originate from hollow carbon atoms sputtered from the surface.

The possibility of using highly charged ions as probes for spin-polarized surfaces is demonstrated in chapter 5. As a first target, a GaAs surface was spin-polarized by optical pumping with polarized laser light. In order to attract the polarized electrons from the bulk to the surface, the surface work function was lowered by adsorption of a small (~ 0.5 ML) amount of cesium atoms onto the surface. Slow He^{2+} ions were scattered off the surface and the projectile KLL Auger emission was used to study the effect of the spin polarization. The changes in the spectral features (peaks) of the Auger spectra were measured as a function of Cs coverage. Maximum change occurred only when σ^+ light was used around a Cs coverage of about 0.5 ML. At this coverage the work function is minimum and the optically pumped and polarized conduction band electrons are expected to be located at the surface. In case the laser was switched off, or in case linearly polarized light was used, no dependence on Cs coverage was found.

As a second target, we used a ferromagnetic nickel surface. This time, the changes in the projectile KLL Auger spectra were studied as a function of target temperature. By increasing the temperature from $T = 20°C$ to $T = 430°C$, *i.e.* through the Curie point of Ni at $354°C$, the relative intensity of the high-spin state decreased. We attributed this change to a loss in the order of the surface electron spins. By destroying the short-range ferromagnetic ordering of the surface, the probability for simultaneous capture of electrons with identical spin orientation decreases and thus the formation of high-spin states is strongly reduced. This is reflected in the Auger spectra, in which decay from low- and high-spin states can be distinguished.

As a third test, spin polarization of surface electrons was induced by an applied magnetic field. Changes in the spin polarization, induced by a switching of the magnetic field, were studied by ECS. We found a good correlation between (changes in) the degree of circular polarization S/I of the fluorescence light and the changes in the magnetic field. Also the sign and magnitude of S/I, as well as the observed asymmetry, are in line with our expectations. From that it can be concluded that these first results are in accordance with the current interpretation.

In chapter 6 a model is introduced which can describe the spectral features in Auger spectra. It is based on a simple 'free atom model' that allows for autoionization (AI rate 10^{14} Hz) of excited projectile states, which are initially populated by their statistic weights. By comparing the model results to experimental data on metallic and semiconductor surfaces, for various scattering conditions, it became clear that the model was incomplete. However, if the excited projectiles are also allowed to decay via resonant ionization (RI rate 10^{15} Hz), the experimental data can be reasonably well described. The difference between the metal and semiconductor Auger spectra were found to be due to the band gap, which blocks the RI process. We also investigated the effects on the projectile velocity component parallel and perpendicular to the surface. Although the parallel component modifies the density of states of the target electrons, almost no effect on the spectral features is observed. This is due to the high RI rate which overwhelms typical AI rates and therefore strongly determines the spectral features. Also effects of 'observation time', determined by the projectiles parallel velocity component, become less pronounced. Furthermore, it seems possible to measure and (roughly) estimate spin ordering effects for spin-polarized targets, as observed via changes in Auger spectra. This was demonstrated by the temperature measurements of the Ni surface, described in chapter 5.

From the results presented in this thesis, it can be concluded that Auger electron spectroscopy and electron capture spectroscopy can be used to probe changes in the spin polarization of surfaces. Since the ion-surface interaction takes place within an area of several tens of nanometers, and lasts only several tens of femtoseconds, the strong electronic interactions take place within a small volume. This enables ions to locally probe the surface electronic structure. Because the interaction area is much smaller than the average magnetic domain size, these methods are also well suited for studies on magnetic surfaces or thin magnetic films.

The description of the complicated interactions, as given in this thesis, is deliberately kept rather simple. However, this more qualitative approach allows a clear and better understanding of the basic phenomena. Of course it is desirable to improve and expand the theoretical and experimental methods. For future experiments with ECS, one can think of using spectral lines of highly excited projectiles. Then it would be possible to measure at elevated target temperatures with no background from black-body radiation. With the aid of density matrix theory [103], it is possible to calculate the polarization of the light for different scattering geometries and different spectral lines. By comparing these with experimental results, one could *e.g.* study the population of initially excited projectile states. Also the model to describe Auger spectra offers room for improvements. For example, it would be interesting to model distance and time dependent RN and RI rates. Or one could *e.g.* try to implement other (real) densities of states and study their influence on the spectral features of the Auger spectra.